

## CLAIMS

What is claimed is:

1. A method of modifying a work-piece surface, the method comprising:
  - (a) introducing a first liquid to a first cluster generation site, the first liquid comprising hydroxylamine or hydroxylamine derivative compound;
  - (b) subjecting the first liquid to electrical forces higher than a surface tension of the first liquid under conditions to prepare a first plurality of clusters; and
  - (c) impacting at least a portion of the clusters of the first plurality of clusters upon a first portion of the surface.
2. The method of claim 1, wherein the oxidizing agent comprises at least one of aqueous hydroxylamine or a hydroxylamine substituted with one or two R groups which can independently be a C<sub>1</sub> to C<sub>4</sub> alkyl and a carboxyl, or salt thereof.
3. The method of claim 2, wherein the first liquid further comprises a solvent.
4. The method of claim 2, wherein, relative to the weight of the first liquid, the total weight of the at least one of hydroxylamine, the derivative of hydroxylamine, the salt of hydroxylamine, and the a salt of a derivative of hydroxylamine is at least 15 percent.
5. The method of claim 4, wherein the oxidizing agent comprises at least one of hydroxylamine and a derivative of hydroxylamine and the total weight thereof is at least 15 percent relative to the weight of the liquid.
6. The method of claim 3, wherein the first liquid consists essentially of the solvent and the at least one of hydroxylamine, the derivative of hydroxylamine, the salt of hydroxylamine, and the a salt of a derivative of hydroxylamine.
7. The method of claim 3, wherein the solvent comprises an organic compound having at least one hydroxyl group.

8. The method of claim 7, wherein the solvent comprises an organic compound having fewer than 20 carbon atoms and at least two hydroxyl groups.

9. The method of claim 1, wherein the first liquid comprises an acid.

10. The method of claim 9, wherein the acid comprises at least one of nitric acid, acetic acid, sulfuric acid, peroxyomonosulfuric acid, perchloric acid, peracetic acid, perchromic acid, periodic acid, perchloric acid, perbromic acid, perfluoric acid, and perboric acid.

11. The method of claim 1, wherein the first liquid comprises at least one corrosion inhibitor.

12. The method of claim 11, wherein the corrosion inhibitor comprises at least one of benzotriazole and a derivative of benzotriazole.

13. A method of modifying a work-piece surface, the method comprising:

- ~ (a) introducing a first liquid to a first cluster generation site, the first liquid comprising periodic acid or hydrogen peroxide;
- ~ (b) subjecting the first liquid to electrical forces higher than a surface tension of the first liquid under conditions to prepare a first plurality of clusters; and
- ~ (c) impacting at least a portion of the clusters of the first plurality of clusters upon a first portion of the surface.

14. The method of claim 13, wherein the total weight of the at least one oxidizer having at least one peroxy group is at least 15 percent relative to the weight of the first liquid.

15. The method of claim 13, wherein the first liquid comprises at least one corrosion inhibitor.

16. The method of claim 13, wherein the corrosion inhibitor comprises at least one of benzotriazole and a derivative of benzotriazole.

17. The method of claim 1, further comprising:

- (d) introducing a second liquid to a second cluster generation site, the second liquid comprising a corrosion inhibitor;
- (e) subjecting the second liquid to electrical forces higher than a surface tension of the second liquid to prepare a second plurality of clusters; and
- (f) impacting clusters of the second plurality clusters upon a second portion of the surface.

18. The method of claim 17, wherein the step of (c) impacting comprises impacting the surface with clusters having a first average kinetic energy and the step of (f) impacting comprises impacting the surface with clusters having a second, smaller average kinetic energy.

19. The method of claim 18, wherein the step of (f) impacting comprises impacting the surface with clusters having an average kinetic energy at least 25% less than an average kinetic energy of the first plurality of clusters.

20. The method of claim 17, wherein the corrosion inhibitor comprises at least one of benzotriazole and a derivative of benzotriazole.

21. The method of claim 17, wherein the corrosion inhibitor comprises at least one of a dihydroxybenzene or trihydroxybenzene.

22. The method of claim 17, wherein at least a portion of the second portion of the surface is also subjected to the step of (c) impacting.

23. The method of claim 22, wherein the step of (f) impacting is performed after initiating the step of (c) impacting.

24. The method of claim 22 wherein, for the portion of the second portion of the surface also subjected to the step of (c) impacting, the step of (f) impacting is performed for a total time T, and further wherein the step of (f) impacting is performed for at least a time  $0.8 \times T$  subsequent to terminating the step of (c) impacting.

25. The method of claim 17, wherein the steps of (c) impacting and (f) impacting are performed while subjecting the first and second portions of the surface to a gas pressure of less than 500 torr and, once one of the steps of (c) impacting and (f) impacting has been initiated, the first and second portions of the surface are not subjected to a gas pressure of greater than 500 torr until at least after the other of the steps of (c) impacting and (f) impacting has also been initiated.

26. The method of claim 17, wherein at least a portion of the first portion of the surface is not subjected to the step of (f) impacting.

27. The method of claim 17, wherein the first and second cluster generation sites are the same.

28. The method of claim 1, wherein the first liquid is essentially free of metals.

29. The method of claim 1, further comprising the steps of:

(g) depositing at least one organometallic compound upon a second portion of the surface; and

(h) annealing the at least one organometallic compound to thereby provide the second portion of the surface with a metallic coating.

30. The method of claim 29, wherein the step of (g) depositing comprises:

(i) introducing a third liquid to a third cluster generation site, the third liquid comprising the at least one organometallic compound;

(j) subjecting the third liquid to electrical forces higher than a surface tension of the third liquid to prepare a third plurality of clusters; and

(k) impacting clusters of the third plurality clusters upon a third portion of the surface.

31. The method of claim 29, wherein the step of (g) depositing comprises depositing at least two different metals upon the second portion of the surface, and wherein the metallic coating provided in the step of (h) annealing comprises at least one alloy.

32. The method of claim 22, wherein the steps of (c) impacting and (k) impacting are performed while subjecting the first and third portions of the surface to a gas pressure of less than 500 torr and, once one of the steps of (c) impacting and (k) impacting has been initiated, the first and second portions of the surface are not subjected to a gas pressure of greater than 500 torr until at least after the other of the steps of (c) impacting and (k) impacting has also been initiated.

33. The method of claim 29, wherein the step of (c) impacting comprises impacting the surface with clusters having a first average kinetic energy and the step of (k) impacting comprises impacting the surface with clusters having a second, smaller average kinetic energy.

34. The method of claim 32, wherein the step of (k) impacting comprises impacting the surface with clusters having an average kinetic energy at least 25% less than an average kinetic energy of the first plurality of clusters.

35. The method of claim 29, wherein at least a portion of the first portion of the surface is also subjected to the step of (k) impacting.

36. The method of claim 35, wherein the step of (k) impacting comprises focusing clusters of the third plurality of clusters.

37. The method of claim 31, wherein, for at least  $1 \times 10^{-5} \text{ cm}^2$  of the third portion of the surface, the step of (g) depositing comprises depositing at least about  $9 \times 10^{-8} \text{ g/cm}^3$  of metal per square centimeter.

38. The method of claim 29, wherein the first and third cluster generation sites are the same.

39. A method for modifying a surface, the method comprising:

(a) introducing a first liquid to a first cluster generation site, the first liquid comprising a reducing agent;

(b) subjecting the first liquid to electrical forces higher than a surface tension of said liquid to prepare a first plurality of clusters; and

(c) impacting clusters of the first plurality of clusters upon a first portion of the surface.

40. The method of claim 39, wherein the reducing agent comprises at least one of hydrazine and a derivative of hydrazine.

41. The method of claim 40, wherein the total weight of the at least one of hydrazine and a derivative of hydrazine is at least 10 percent relative to the weight of the first liquid.

42. The method of claim 40, wherein the first liquid is essentially free of metals.

43. The method of claim 40, further comprising:

(d) introducing a second liquid to a second cluster generation site, the second liquid comprising a corrosion inhibitor;

(e) subjecting the second liquid to electrical forces higher than a surface tension of the second liquid to prepare a second plurality of clusters; and

(f) impacting clusters of the second plurality clusters upon the surface.

44. The method of claim 43, wherein the corrosion inhibitor comprises at least one of benzotriazole and a derivative of benzotriazole.

45. A method for modifying a surface, the method comprising:

(a) introducing a first liquid to a first cluster generation site, the first liquid comprising an organometallic compound;

(b) subjecting the first liquid to electrical forces higher than a surface tension of the first liquid to prepare a first plurality of clusters; and

(c) impacting clusters of the first plurality of clusters upon the surface, wherein at least some of the organometallic compound deposits upon the surface.

46. The method of claim 45, further comprising the step of:

(d) subjecting the surface deposited organometallic compound to annealing whereby an at least partial metallic coating is formed upon the surface, wherein the step of (c)

impacting comprises depositing at least two different metals upon the surface and the step of (d) subjecting comprises formation of a metallic alloy upon the surface.